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THERMODYNAMIC FUNCTIONS OF GAS MIXTURES AT HIGH TEMPERATURES

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ABSTRACT

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Needs for simple, accurate analytical expressions of gas properties at high temperatures and uses are discussed. The problem of pure and mixed gases and other specific drawbacks in referenced works are discussed. Expressions used by Zdunkevich in his computer work and accuracy of the 50-card program are discussed.

Author

For the investigation of a large number of modern scientific and technical problems it is necessary to have simple and sufficiently accurate analytical expressions for the thermodynamic, transport and physical properties of gases and gas mixtures, applicable from several degrees to 15000 - 20000° K. Such analytical expressions could be used, for example, in compiling standard computer programs and during solution of various problems of aeromechanics and plasma physics, since the volume of the memory of the existing computers does not permit the use of appropriate tables even if they are available. The analytical expressions for thermodynamic functions of pure monoatomic or diatomic gas, the accuracy of which is 0.5 percent in the range from 10 to 15000 - 20000° K and pressures up to 10^7 - 10^8 N/m^2, were obtained in reference 1.

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*Numbers given in the margin indicate the pagination in the original foreign text.

In practice, however, it is frequently necessary to have analytical expressions not for pure gas, but for a gas mixture such as air. A small number of the existing works on the analytical description of thermodynamic functions of air may be subdivided into two groups. The first group (refs. 2, 3) formally approximates the available data from the tables (ref. 4) the calculation of which for the determination of the composition of air involves solution of a complex system of nonlinear algebraic equations, including the mass action expressions for all possible reactions in air, Dalton's law, conservation of mass for each of the components, and conservation of charge. However, the complex nature of the temperature and pressure dependence of the thermodynamic functions necessitates the availability of different approximation formulas or coefficients for different intervals. This significantly lessens the value of these formulas.

The second group of works (refs. 5 - 8) is of great interest since here the real physical processes which occur in air are considered in the first approximation. V. V. Mikhaylov (ref. 5) used the "ideally dissociating gas" model proposed by M. Leightkill (ref. 6) to a mixture of nitrogen and oxygen and obtained relatively simple expressions for the thermodynamic functions for air in the 2000 - 15000 K and 10^2 - 10^8 N/m^2 region. The deficiency of this work is its low accuracy, caused by the crudeness of many assumptions, the major ones of which are: 1-dissociation of molecular nitrogen and oxygen is calculated for the "ideally dissociating gas" model; 2-the formation of nitric oxide is not included; 3-it is assumed that all chemical reactions proceed independently; 4-molecular vibrations are not included; 5-argon is not included in the calculations.

The second drawback is that a number of constants which are used in this work may be selected only from accurate table data, consequently the analytical

expressions which are given in reference 5 are completely inapplicable for calculation of the thermodynamic functions of random mixtures of nitrogen, oxygen and argon. F. Hansen (ref. 7) obtained a somewhat better approximation than V. V. Mikhaylov primarily because he used a more accurate composition of air. However, the use of a large number of terms of statistical sums of various components and the necessity of numerical differentiation and integration to obtain such quantities as c_p, c_v, S and a, significantly complicates the program and lowers the speed of computer calculations and assumptions 2 - 5, which are also used in this work, do not significantly improve the accuracy. A good agreement with the table data can be obtained only by further complex processing of the results of V. V. Mikhaylov or F. Hansen by means of formal approximation of deviations, as this was done in the work of I. N. Naumova (ref. 8).

On the basis of earlier published accurate analytical expressions for pure gas (ref. 1) which we obtained in this work, simple analytical expressions for the composition and thermodynamic functions of random mixtures of nitrogen, oxygen and argon (and consequently air) take into account the unharmoniousness of /640
molecular vibrations, electric excitation of particles, reactions involved in the formation of nitric oxide, equilibrium dissociation and ionization. Since in this work we do not make the assumptions which were made by V. V. Mikhaylov and F. Hansen, the relative error in calculating any of the thermodynamic functions is much smaller than in references 5 - 7.

The analytical expressions which are given in this work were used by M. D. Sdunkevich in compilation of a computer program for calculation of the thermodynamic functions for random mixtures of nitrogen, oxygen and argon in a braod temperature interval (up to the second ionization stage). This program adjusted was used for the calculation of the thermodynamic functions of nitrogen as

well as for a mixture of nitrogen (98.5 percent by volume under normal conditions), argon (1.2 percent) and oxygen (0.3 percent) which is the atmosphere of Mars (according to the 1963 data), in the temperature range from 100 to 12000 - 20000° K and pressures from 10^{-1} to 10^{8} N/m^2. Here the maximum relative error for nitrogen as compared with earlier published data (ref. 9) does not exceed 0.5 percent and for air -- 1 percent.

1. Equation of state and equilibrium composition of gas mixtures at high temperatures. It is well known that thermodynamic functions of gas mixtures are determined by the thermodynamic parameters of all their components and composition which changes with a change of temperature and pressure. Although from the theoretical standpoint the calculation of the composition of the mixture is possible, yet the direct solution of an appropriate system of nonlinear algebraic equations for the case of a large number of reactions, many of which proceed simultaneously, leads to cumbersome calculations. Thus, in accurate determination of the composition of air in the 6000 - 12000° K temperature interval, the complete system of chemical equilibrium equations includes equations for mass action for each of the ten reactions which are considered for air, Dalton's law, material, balance equation and conservation of charge equations (ref. 10). It is obvious that such a system of equations may be solved only by numerical methods on a computer.

In this work, during the determination of the composition of a random mixture of nitrogen, oxygen and argon, we limit ourselves to consideration of the following basic six chemical reactions which may be broken down into three groups:

1) nitric oxide formation reaction

$$N_2 + O_2 \rightleftarrows 2NO + E_{NO}$$

2) dissociation of molecular nitrogen and oxygen

$$A_{2j} \rightleftarrows 2A_{1j} + D_j,$$

3) ionization of atomic compounds

$$A_{1j} \rightleftarrows A_{1j} + e + I_j \quad (j = O, N, Ar).$$

Resulting from all the other reactions (formation of higher oxides of nitrogen, dissociation of NO, ionization of molecular components, etc.) components are produced whose concentrations do not exceed 0.1 percent in terms of the order of magnitude.

Let us consider some unit volume of a mixture of nitrogen, oxygen and argon, taking into account the above-indicated basic chemical reactions. Let us consider that the mixture exists in a state of complete thermodynamic equilibrium and consequently is characterized not only by pressure, but also by temperature and composition. Let the investigated volume contain $n_{2j}(j = O, N)$ number of diatomic molecules, of which n_{NO} has reacted forming nitric oxide, and thus the mixture contains $2n_{NO}$ molecules of NO. Let the mixture also contain $2n_{ij}(j = O, N, Ar)$ atoms, of which $2n_{ij}$ are singly ionized, and thus the mixture contains /641 also $\sum_j 2n_{ij}$ free electrons.

The density of gas in the considered volume may be written as follows

$$\rho = \sum_{k=1}^{4} \sum_{j=O,N,Ar} m_{kj} n_{kj} + m_{NO}\, 2n_{NO} = \sum_j [m_{2j}(n_{2j} - n_{NO}) + m_{1j}(2n_{1j} - 2n_{ij}) + \\ + m_{ij} 2n_{ij} + m_e 2n_{ij}] + m_{NO} 2n_{NO} = \sum_j m_{2j} (n_{2j} - n_{NO} + n_{1j}) + m_{NO} 2n_{NO}, \\ (m_{2j} = 2m_{1j} = 2 (m_{ij} + m_e). \tag{1.1}$$

Here index k designates one of four considered states of the j-th component (k=2- diatomic molecule; 1-atom; i- ion; e- electron), where for argon $(n_{2Ar} - n_{NO}) \equiv 0$.

The degree of dissociation $\bar{\alpha}_j$ of the j-th component in the mixture may be determined as the ratio of the dissociated mass of the j-th component to the total mass of this component in the mixture, i.e.,

$$\bar{\alpha}_j = \frac{m_{1j} 2n_{1j}}{m_{2j}(n_{2j} - n_{NO} + n_{1j})} = \frac{n_{1j}}{n_{2j} - n_{NO} + n_{1j}}, \tag{1.2}$$

where according to definition $\bar{\alpha}_{Ar} \equiv 1.0$.

Analogously, $\bar{\beta}_j$ is the degree of ionization of the j-th component in the mixture, defined as the ratio of the mass of charged particles formed as a result of the ionization of the j-th atomic component to the total mass of this atomic component

(1.3)

Let us now derive an equation of state for the mixture, taking into account the chemical reactions. According to Dalton's law the total pressure of the mixture in this case may be written as follows

$$P = kT\left[\sum_{k=1}^{4}\sum_{j=0,N,Ar}^{j} n_{kj} + 2n_{NO}\right] = kT\left[\sum_{j}(n_{2j} - n_{NO} + 2n_{1j} + 2n_{ij}) + 2n_{NO}\right].$$

Taking into account (1.1) and also the conservation of mass equation for the j-th component

$$\frac{n_{2j} - n_{NO} + n_{1j}}{n_{2i} - n_{NO} + n_{1i}} = \frac{x_j}{x_i},$$

and the equation of state of this mixture of gases is finally written as follows

$$Z = \frac{P}{\rho R_0 T} = \frac{\sum_j x_j [1 + \bar{\alpha}_j (1 + 2\beta_j)] + x_{NO}}{1 + x_{NO}}, \tag{1.4}$$

where Z is the compressibility; $R_0 = R_{un}/\mu_0$; x_j and $\mu_0 = 2\sum_j m_{1j} x_j$ are constants /642

which characterize the volume composition and molecular weight of the mixture under normal conditions; $x_{NO}=x_{NO}(x_j, T, P)$--the volume concentration of NO.

To calculate the compressibility and other thermodynamic functions it is necessary to know the composition of the mixture at elevated temperatures, i.e., parameters $\bar{\alpha}_j$, $\bar{\beta}_j$ and x_{NO}. Considering the reaction of formation of NO, on the basis of the mass action law we may write

$$x_{NO} = c_{NO}\sqrt{x_O x_N}\sqrt{(1-\bar{\alpha}_O)(1-\bar{\alpha}_N)}\exp\left(-\frac{E_{NO}}{kT}\right), \tag{1.5}$$

where c_{NO} and E_{NO} are constants. From (1.5) it is apparent that dissociation and ionization reactions of NO need not be considered since almost always the dissociation of oxygen terminates (i.e., $\alpha_O \approx 1.0$) by the beginning of dissc $\bar{\beta}_j = \frac{m_{ij}^2 n_{ij} + m_e^2 n_{ij}}{m_{1j}^2 n_{1j}} = \frac{n_{ij}}{n_{1j}}$. or ionization of NO.

During calculation of $\bar{\alpha}_j$, the degree of dissociation of the j-th component in the mixture, we shall take an appropriate quantity α_j for the pure gas known from reference 1 and correct it for the effect of the chemical reaction on the partial pressure of the considered component. Here the relationship of α_j to α_j may be written as follows

$$\bar{\alpha}_j = -\frac{b_j}{2}\alpha_j^2 + \sqrt{(1+b_j)\alpha_j^2 + \left(\frac{b_j}{2}\alpha_j^2\right)^2}, \tag{1.6}$$

where for oxygen and nitrogen

$$b_O = \frac{x_N(1+\bar{\alpha}_N) + 2x_{Ar} + x_{NO}}{x_O(1+x_{NO})}, \qquad b_N = \frac{x_O(1+\bar{\alpha}_O) + 2x_{Ar} + x_{NO}}{x_N(1+x_{NO})}. \tag{1.7}$$

In the case of pure gas $b_j \equiv 0$ and $\bar{\alpha}_j \equiv \alpha_j$. The system of equations (1.6), (1.7) is solved by the successive approximation method. For mixtures in which nitrogen is predominant, as the first approximation, one may assume $\bar{\alpha}_N \approx \alpha_N$ and $x_{NO} = 0$ (this approximation is generally sufficient for pressures less than

10^5 N/m^2). Then we calculate b_o and subsequently $\bar{\alpha}_o$, and finally x_{NO} and $\bar{\alpha}_N$. The number of approximations depends primarily on the pressure of the gas. For air at pressures from 10^5 to 10^8 N/m^2 ;it is sufficient to use two approximations and for the mixture which comprises the atmosphere of Mars, due to very low concentration of molecular oxygen, the NO formation reaction need not be considered at all, while dissociation reactions of oxygen and nitrogen are considered separately.

During the calculation of the degree of ionization $\bar{\beta}_j$, let us take into consideration the fact that the corresponding magnitudes of β_j for pure gases, as a rule, differ insignificantly from each other, and consideration of the partial pressures of these components in such mixtures as air or the atmosphere of Mars, where the nitrogen is predominant, even further decreases the small differences in β_j. Thus, the ionization of mixtures of nitrogen, oxygen and argon may be treated as ionization of a single "pure" atomic component, possessing the averaged properties of nitrogen, oxygen and argon atoms, taking into account the ratio of their volumes. Mathematically this is written as follows

$$\left(T_i = \frac{\Sigma x_j T_{ij}}{\Sigma x_j},\ \theta_A = \frac{T_A^E}{T},\ \theta_i = \frac{T_i^E}{T},\quad c_i \frac{1 + (g_A e^{\theta_A})^{-1}}{1 + (g_i e^{\theta_i})^{-1}} = c_{i_0} \frac{\Sigma x_j Z_{Aj}}{\Sigma x_j Z_{ij}}\right),$$

$$\bar{\beta} = \bar{\beta}_j = \left\{1 + \frac{c_i P}{T^{5/2}} \frac{\sum_j x_j (1 + \bar{\alpha}_j)}{2 \sum_j x_j \bar{\alpha}_j} \exp\left(\frac{\bar{T}_i}{T}\right) \frac{1 + (g_A e^{\theta_A})^{-1}}{1 + (g_i e^{\theta_i})^{-1}}\right\}^{-\frac{1}{2}} \qquad (1.8)$$

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where x_j, T_{ij}, g_A, g_i, T_i, c_i, c_{i_o}, T_A^E and T_i^E are constants.

2. Thermodynamic functions of gas mixtures. The specific internal energy of a gas mixture is equal to the sum of the energies of the individual components and it is written as follows

$$u = \frac{R_0}{1 + x_{NO}} T \left\{\sum_j x_j \varepsilon_j + x_{NO}\left(\frac{T_{NO}}{T} + \frac{5}{2} + \varepsilon_{v,NO}\right)\right\} + u_0, \qquad (2.1)$$

where

$$\varepsilon_j = \frac{5+\bar{\alpha}_j}{2} + 3\bar{\alpha}_j\bar{\beta} + (1-\bar{\alpha}_j)\,\varepsilon_{vj} + 2\bar{\alpha}_j(1-\bar{\beta})\,\varepsilon_{Aj} + 2\bar{\alpha}_j\bar{\beta}\varepsilon_{ij} + \bar{\alpha}_j\frac{T_{Dj}}{T} + 2\bar{\alpha}_j\bar{\beta}\,\frac{T_{ij}}{T},$$

and the explicit expressions for ε_{vj}, ε_{Aj} and ε_{ij} may be found in reference 1.

Since the equilibrium composition, the internal energy, and the equation of state of the reacting gas mixture are known, the calculation of the remaining thermodynamic functions in principle does not present any difficulties. Thus, in the considered case the specific enthalpy of the mixture is equal to

$$h = u + \frac{P}{\rho} = \frac{R_0}{1+x_{NO}}\,T\left\{\sum x_j\,[\varepsilon_j + 1 + \bar{\alpha}_j(1+2\bar{\beta})] + x_{NO}\left(\frac{T_{NO}}{T} + \frac{7}{2} + \varepsilon_{v\,NO}\right)\right\} + u_0, \tag{2.2}$$

where for argon $\varepsilon_{Ar} = 2\,\bar{\beta}\,(T_{\bar{i}}/T+3+\varepsilon_i)$, since $\alpha_{Ar}\equiv 1.0$ and $D_{Ar}=0$.

Omitting the intermediate steps we shall write the final expression for the remaining thermodynamic functions. For the ratio of specific heats we have

$$\varkappa = \frac{\sum x_j\,[7 + 3\bar{\alpha}_j + 10\bar{\alpha}_j\bar{\beta} + 2(1-\bar{\alpha}_j)\,c_{vj} + 4\bar{\alpha}_j(1-\bar{\beta})\,c_{Aj} + 4\bar{\alpha}_j\bar{\beta}c_{ij} + \Delta c_{pj}] + 2x_{NO}\left(\frac{7}{2} + c_{v\,NO} + \Delta c_{p\,NO}\right)}{\sum x_j\,[5 + \bar{\alpha}_j + 6\bar{\alpha}_j\bar{\beta} + 2(1-\bar{\alpha}_j)\,c_{vj} + 4\bar{\alpha}_j(1-\bar{\beta})\,c_{Aj} + 4\bar{\alpha}_j\bar{\beta}c_{ij} + \Delta c_{vj}] + 2x_{NO}\left(\frac{5}{2} + c_{v\,NO} + \Delta c_{v\,NO}\right)}, \tag{2.3}$$

where

$$\Delta c_{pj} = \frac{\alpha_j^2(1-\alpha_j^2)}{\bar{\alpha}_j}\,\frac{1+b_j(1-\bar{\alpha}_j)}{1+\frac{b_j}{2}\frac{\alpha_j^2}{\bar{\alpha}_j}}\,A_j\,(A_j + 2\bar{\beta}B) + 2\bar{\alpha}_j\bar{\beta}\,(1-\bar{\beta}^2)\,B^2,$$

$$\Delta c_{vj} = \frac{2\bar{\alpha}_j(1-\bar{\alpha}_j)}{2-\bar{\alpha}_j}(A_j-1)\,[A_j - 1 + 2\bar{\beta}\,(B-1)] + \bar{\alpha}_j\frac{4\bar{\beta}(1-\bar{\beta})}{2-\bar{\beta}}(B-1)^2,$$

$$\Delta c_{p\,NO} = \frac{1}{1+x_{NO}}\left(\frac{1}{2}\frac{T_{NO}}{T} - \sum x_j\bar{\alpha}_jA_j\right)\left[\frac{T_{NO}}{T} - \frac{1}{2}\sum x_j\frac{\Delta c_{pj}}{(1-\bar{\alpha}_j)\,(A_j+2\bar{\beta}B)}\right],$$

$$\Delta c_{v\,NO} = \frac{1}{1+x_{NO}}\left[\frac{T_{NO}}{2T} - \sum x_j\bar{\alpha}_j\,(A_j-1)\right]\left\{\frac{T_{NO}}{T} - \frac{1}{2}\sum x_j\frac{\Delta c_{vj}}{(1-\bar{\alpha}_j)[A_j-1+2\bar{\beta}(B-1)]}\right\},$$

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The equilibrium velocity of sound in the reacting medium is calculated by the following formula

$$a^2 = c_a^2 \frac{\varkappa T}{1+\delta} \frac{\sum x_j [1 + \bar{\alpha}_j (1 + 2\bar{\beta})] + x_{NO}}{1 + x_{NO}}, \tag{2.4}$$

where c_a is constant and

$$\delta = \frac{\sum x_j \left[\frac{\alpha_j^2}{\alpha_j} (1 - \alpha_j^2)(1 + 2\bar{\beta}) \frac{1 + b_j (1 - \bar{\alpha}_j)}{1 + b_j \alpha_j^2 / 2\bar{\alpha}_j} + 2\bar{\alpha}_j \bar{\beta} (1 - \bar{\beta}^2) \right] + \frac{2x_{NO}}{1 + x_{NO}}}{2 \left\{ \sum x_j [1 + \bar{\alpha}_j (1 + 2\bar{\beta})] + x_{NO} \right\}},$$

$$\frac{S - S_0}{R_0} = \left\{ \sum x_j \left[\frac{7 + 3\bar{\alpha}_j + 10\bar{\alpha}_j \bar{\beta}}{2} \right] \right\} \ln T -$$

$$- \left\{ \sum x_j [1 + \bar{\alpha}_j (1 + 2\bar{\beta})] \right\} \ln P + \sum x_j S_j + x_{NO} S_{NO}^{(1)}, \tag{E}$$

and where

$$S_j = (1 - \bar{\alpha}_j) S_{vj} + 2\bar{\alpha}_j (1 - \bar{\beta}) S_{Aj} + 2\bar{\alpha}_j \bar{\beta} S_{ij} + \bar{\alpha}_j S_j^{(1)} -$$
$$- 2\bar{\alpha}_j \bar{\beta} S_j^{(2)} + (1 + b_j + \bar{\alpha}_j) \ln (1 + b_j + \bar{\alpha}_j) - (1 + b_j) \ln (1 + b_j) -$$
$$- (1 - \bar{\alpha}_j) \ln (1 - \bar{\alpha}_j) - 2\bar{\alpha}_j \ln 2\bar{\alpha}_j + 2\bar{\alpha}_j [(1 + \bar{\beta}) \ln (1 + \bar{\beta}) -$$
$$- (1 - \bar{\beta}) \ln (1 - \bar{\beta}) - 2\bar{\beta} \ln 2\bar{\beta}], \tag{2.5}$$

Here $S_j^{(1)}$ and $S_j^{(2)}$ are constants.

It also follows from (2.5) that the adiabatic flow of the mixture, when all chemical reactions and even vibrations are "frozen," is calculated from the well-known relationship for the perfect gas with the following heat capacity ratio

$$\varkappa^0 = \frac{\sum x_j [7 + 3\bar{\alpha}_j^0 + 10\bar{\alpha}_j^0 \bar{\beta}^0] + 7x_{NO}^0}{\sum x_j [5 + \bar{\alpha}_j^0 + 6\bar{\alpha}_j^0 \bar{\beta}^0] + 5x_{NO}^0}, \tag{2.6}$$

where the superscript "nill" designates a constant, "frozen" value of the composition of the mixture.

The presented analytical expressions were utilized by M. D. Zdunkevich during the compilation of a computer program for the calculation of thermodynamic properties (μ, h, u, S, c_p, c_v, a) transport properties (η, λ, Pr, Le) and some physical properties (Debyé radius r_D and conductivity σ) of a random mixture of

nitrogen, oxygen and argon in a broad pressure and temperature range (up to the second ionization stage).

The whole program consists of 50 punched cards. The program was adjusted for the calculation of the indicated thermodynamic, transport and physical properties of nitrogen, oxygen and air in the temperature range from a few degrees to 12000 - 20000° K (depending on the pressure) and at pressures from 10^{-1} to 10^{8} N/m^2. The comparison of calculations according to this program with the available table data indicates that the error in calculation of the thermo- /645
dynamic functions for pure gases does not exceed 0.5 percent and for air it does not exceed 1 percent.

The sample of the accuracy of calculations for air is shown in the table which contains the accurate values of μ, h and S for air pressure $P=1.01325 \cdot 10^5$ N/m^2 (ref. 4) and the magnitude of deviations of the calculated values by the developed program from the true values. For comparison there are given the corresponding deviations for the approximations of V. V. Mikhaylov (ref. 5). It is necessary to note that at the present time V. V. Mikhaylov compiled a program for the calculation of thermodynamic functions (μ, h, a) for mixtures of nitrogen, oxygen and carbon dioxide, having the same accuracy as shown in ref. 5.

During calculations of gas dynamic functions, entropy changes the relative error $(e^{\Delta S/Ro} - 1) \cdot 100$ percent, where for air $R_o = 0.0686$.

The error in the calculation of the transport properties does not exceed 5 - 10 percent for pure gases and 10 - 20 percent for air, which is sufficiently small for this type of calculation.

Table 1

T°K	1000	2000	3000	4000	5000	6000	8000	10000	12 000
μ [2]	28.97	28.96	28.31	25.18	23.91	22.04	15.39	14.25	13.03
$\Delta\mu$ [5]	0	0	—0.05	—0.40	—0.20	—0.05	—0.05	—0.1	—0.15
$\Delta\mu$	0	0	0	—0.01	0.01	0	0	0.01	0.01
h [2]	250.0	544.6	972.4	1832	2448	3573	9024	11410	14880
Δh [5]	25	10	—35	5	—15	—20	—140	—400	—650
Δh	—0.2	0.1	0.8	—3	—6	—10	8	10	—50
S [2]	2.06	2.262	2.432	2.678	2.816	3.018	3.799	4.066	4.366
ΔS [5]			~ —0.025			0.005	—0.020	—0.050	—0.070
ΔS	0	—0.002	—0.003	—0.001	0	0	0	—0.001	—0.003

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